

**TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371**

022701-951

U.S. APPLICATION NO. (If known, see 37 C.F.R. 1.5)

UNASSIGNED

09/936895

INTERNATIONAL APPLICATION NO.  
PCT/FR00/00687INTERNATIONAL FILING DATE  
20 MARCH 2000PRIORITY DATE CLAIMED  
19 MARCH 1999

## TITLE OF INVENTION

USE OF COPOLYMERS DERIVED FROM POLYAMIDES AS GELLING AGENTS FOR COMPOUNDS WITH LOW POLARITY

## APPLICANT(S) FOR DO/EO/US

Edith CANIVENC et al.

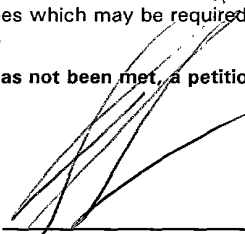
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and the PCT Articles 22 and 39(1).
4. ☐ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ has been transmitted by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
- ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
- ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☒ have not been made and will not be made.
- ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

## Items 11. to 16. below concern other document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.  
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:

Official Notification from the I.B. of the WIPO (Form PCT/IB/308); Official Notification of Election from the I.B. (Form PCT/IB/332); International Search Report; Published Application No. WO 00/56852.

U.S. APPLICATION NO. (if known) <b>09/936895</b> <b>UNASSIGNED</b>		INTERNATIONAL APPLICATION NO. <b>PCT/FR00/00687</b>		ATTORNEY'S DOCKET NUMBER <b>022701-951</b>	
17. <input checked="" type="checkbox"/> The following fees are submitted:				<b>CALCULATIONS</b>	PTO USE ONLY
<b>Basic National Fee (37 CFR 1.492(a)(1)-(5)):</b>  Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... \$1,000.00 (960)  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... \$860.00 (970)  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$710.00 (958)  International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$690.00 (956)  International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$100.00 (962)					
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>					
Surcharge of <b>\$130.00 (154)</b> for furnishing the oath or declaration later than 20 <input type="checkbox"/> 30 <input type="checkbox"/> months from the earliest claimed priority date (37 CFR 1.492(e)).				\$ 860.00	
Claims	Number Filed	Number Extra	Rate		
Total Claims	23 -20 =	3	X\$18.00 (966)	\$ 54.00	
Independent Claims	1 -3 =	0	X\$80.00 (964)	\$ 0.00	
Multiple dependent claim(s) (if applicable)			+ \$270.00 (968)	\$ 0.00	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$ 54.00	
Reduction for 1/2 for filing by small entity, if applicable (see below).				\$	-
<b>SUBTOTAL =</b>				\$ 914.00	
Processing fee of <b>\$130.00 (156)</b> for furnishing the English translation later than 20 <input type="checkbox"/> 30 <input type="checkbox"/> months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
<b>TOTAL NATIONAL FEE =</b>				\$ 914.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). <b>\$40.00 (581)</b> per property +				\$	
<b>TOTAL FEES ENCLOSED =</b>				\$ 914.00	
				<b>Amount to be:</b>	
				<b>refunded</b>	\$
				<b>charged</b>	\$
a. <input type="checkbox"/> Small entity status is hereby claimed. b. <input checked="" type="checkbox"/> A check in the amount of \$ <u>914.00</u> to cover the above fees is enclosed. c. <input type="checkbox"/> Please charge my Deposit Account No. <u>02-4800</u> in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed. d. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>02-4800</u> . A duplicate copy of this sheet is enclosed.					
<b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</b>					
SEND ALL CORRESPONDENCE TO:					
Teresa Stanek Rea BURNS, DOANE, SWECKER & MATHIS, L.L.P. P.O. Box 1404 Alexandria, Virginia 22313-1404 (703) 836-6620					
 _____ SIGNATURE					
_____ NAME					
30,427 REGISTRATION NUMBER					

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Patent Application of	)	
Edith CANIVENC et al.	)	
	)	Group Art Unit: (Unassigned)
Application No.: Unassigned	)	
(Corresponds to PCT/FR00/00687)	)	Examiner: (Unassigned)
	)	
International Filing Date: March 20, 2000	)	
	)	
For: USE OF COPOLYMERS DERIVED	)	
FROM POLYAMIDES AS GELLING	)	
AGENTS FOR COMPOUNDS WITH	)	
LOW POLARITY	)	

**PRELIMINARY AMENDMENT****BOX PCT**

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

Prior to examination, please amend the above-captioned application as follows:

**IN THE CLAIMS:**

Kindly amend claims 1-23 as follows:

1. (Amended) A gelling agent comprising a relatively nonpolar compound or a nonionic surfactant, comprising at least one block copolymer comprising the following units:

- |                                                              |               |
|--------------------------------------------------------------|---------------|
| -X-(R <sup>1</sup> ) <sub>m</sub> -(OA) <sub>n</sub> -X-     | (I), and      |
| -[NH-R <sup>2</sup> -NHCO-R <sup>2</sup> -CO] <sub>f</sub> - | (IIa), and/or |
| -[NH-R <sup>3</sup> -CO] <sub>s</sub> -                      | (IIb), and    |
| -OC-R <sup>4</sup> -CO-                                      | (III)         |

in which units:

- ☐ X corresponds to the end function of said unit after reaction with an amine or carboxylic acid function;
- ☐  $R^1$ , which may be identical or different, represents a linear or branched alkyl radical comprising 2 to 4 carbon atoms;
- ☐ OA, which may be identical or different, represents an oxyethylenated, oxypropylenated or oxybutylenated radical, or mixtures thereof;
- ☐  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, represent:
  - a linear or branched alkyl radical comprising 2 to 18 carbon atoms,
  - an aryl radical comprising one or more optionally substituted aromatic nuclei,
- ☐ m is equal to 0 or 1,
- ☐ n is an integer between 4 and 800,
- ☐ r is such that the number of amide bonds is between 1 and 15 per unit (I),
- ☐ s is such that the number of amide bonds is between 1 and 15 per unit (I),
- ☐ the molar ratio of the number of units (III) to the number of units (I) is between 0.5/1 and 1.4/1;
- ☐ the various units are linked together at least by means of amide bonds; and
- ☐ the number-average molar mass of the copolymer is less than 100 000 g/mol.

2. (Amended) The gelling agent as claimed in claim 1, wherein the units (I) of the copolymer comprise a sequence of oxyethylenated, oxypropylenated or oxyethylenated/oxypropylenated radicals, the sum of the units being equal to n.

3. (Amended) The gelling agent as claimed in claim 1, wherein the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, represent linear or branched radicals comprising 2 to 12 carbon atoms.

4. (Amended) The gelling agent as claimed in claim 1, wherein the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, are selected from the group consisting of:

- ☐ ethyl, 1-methylethyl, propyl, 1-methylpropyl, butyl, hexyl, heptyl, octyl, decyl, undecyl and lauryl radicals.

5. (Amended) The gelling agent as claimed in claim 1, wherein the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, represent aryl radicals comprising one or more optionally substituted aromatic nuclei.

6. (Amended) The gelling agent as claimed in claim 1, wherein the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, comprise:

- ★ an aromatic nucleus, the reactive functions (amines or carboxylic acids) being in an ortho, meta or para position,
- ★ two aromatic nuclei, linked via inert groups, or peri-fused, the reactive functions (amines or carboxylic acids) being on the carbon atoms 1 and 2, 1 and 4, 1 and 5, 1 and 6, 1 and 7 or 2 and 7.

7. (Amended) The gelling agent as claimed in claim 1, wherein the units (IIa) or (IIb) are selected from the group consisting of polyamide 6, polyamide 10, polyamide 11, polyamide 12, polyamide 6, 6 or a random copolymer of at least two such polyamides, in all proportions.

8. (Amended) The gelling agent as claimed in claim 1, wherein r and s, which may be identical or different, are such that the number of amide bonds is between 5 and 10 per unit (I).

9. (Amended) The gelling agent as claimed in claim 1, wherein the number-average molecular mass of the copolymer is between 10 000 and 50 000 g/mol.

10. (Amended) The gelling agent as claimed in claim 1, wherein the relatively nonpolar compound is in the Hansen solubility space, and has the following parameters:

- $\delta P$  of Keesom interactions of less than or equal to  $16.5 \text{ (J/cm}^3\text{)}^{1/2}$
- $\delta H$  of hydrogen bonds of less than or equal to  $10.5 \text{ (J/cm}^3\text{)}^{1/2}$
- $\delta D$  of London interactions of greater than or equal to  $15 \text{ (J/cm}^3\text{)}^{1/2}$ .

11. The gelling agent as claimed in claim 1, wherein the nonionic surfactant is selected from the group consisting of:

- polyoxyalkylenated (polyethoxyethylenated, polyoxypropylenated or polyoxybutylenated) alkylphenols in which the alkyl substituent is C<sub>6</sub>-C<sub>12</sub> and containing from 5 to 25 oxyalkylene units;
- polyoxyalkylenated C<sub>8</sub>-C<sub>22</sub> aliphatic alcohols containing from 1 to 25 oxyalkylene (oxyethylene or oxypropylene) units;
- products resulting from the condensation of ethylene oxide and/or propylene oxide with propylene glycol or ethylene glycol;
- ethoxylated and/or propoxylated C<sub>8</sub>-C<sub>18</sub> fatty acids containing from 5 to 25 ethoxylated and/or propoxylated units;
- alkoxyated amido amines containing from 1 to 50 oxyalkylenated units;
- alkoxyated terpenic hydrocarbons, containing from 1 to 30 oxyethylene and/or oxypropylene units;
- alkylpolyglycosides which may be obtained by condensation of glucose with primary fatty alcohols containing a C<sub>4</sub>-C<sub>20</sub> alkyl group and also an average number of glucose units of about from 0.5 to 3 per mole of alkylpolyglycoside.

12. (Amended) The gelling agent as claimed in claim 1, wherein the amount of copolymer relative to the relatively nonpolar compound or the nonionic surfactant is between 0.1 % and 15% by weight.

13. (Amended) A method for gelation of a relatively nonpolar compound, comprising using the gelling agent as claimed in claim 1, wherein the copolymer is combined with a nonionic or anionic surfactant.

14. (Amended) The method as claimed in claim 13, wherein the amount of nonionic or anionic surfactant relative to the relatively nonpolar compound is between 5% and 20% by weight.

15. (Amended) The gelling agent as claimed in claim 1, wherein the copolymer is combined with a filler of lamellar structure.

16. (Amended) The gelling agent as claimed in claim 15, wherein the amount of filler represents up to 20% by weight of the copolymer.

17. (Amended) The gelling agent as claimed in claim 15, wherein the filler is introduced during the preparation of the copolymer and/or during the use of said copolymer.

18. (Amended) A composition of formulations intended for cleaning metals comprising an effective amount of the gelling agent according to claim 1.



19. (Amended) A detergent formulation used in the industrial field comprising an effective amount of the gelling agent according to claim 1.

20. (Amended) A formulation intended for stripping paints and varnishes comprising an effective amount of the gelling agent according to claim 1.

21. (Amended) A method intended for cleaning or stripping vertical surfaces comprising using the gelling agent according to claim 15.

22. (Amended) A method for treating plants comprising using an effective amount of the gelling agent according to claim 1.

23. (Amended) A composition for ink printing comprising an effective amount of the gelling agent according to claim 1.

**REMARKS**

Entry of the foregoing amendment(s) is respectfully requested.

The claims have been amended to eliminate multiple dependency and to place them in better condition for U.S. patent practice.

Should the Examiner have any questions concerning the subject application, a telephone call to the undersigned would be appreciated.

Respectfully submitted,

BURNS, DOANE, SWECKER & MATHIS, L.L.P.

By: 

Teresa Stanek Rea  
Registration No. 30,427

P.O. Box 1404  
Alexandria, Virginia 22313-1404  
(703) 836-6620

**Filed: September 19, 2001**

**Attachment to Preliminary Amendment dated September 19, 2001**

**Marked-up Claims 1-23**

1. (Amended) [The use as a] A gelling agent[, of] comprising a relatively nonpolar compound or a nonionic surfactant, [of] comprising at least one block copolymer comprising the following units:

- |                                                              |               |
|--------------------------------------------------------------|---------------|
| -X-(R <sup>1</sup> ) <sub>m</sub> -(OA) <sub>n</sub> -X-     | (I), and      |
| -[NH-R <sup>2</sup> -NHCO-R <sup>2</sup> -CO] <sub>r</sub> - | (IIa), and/or |
| -[NH-R <sup>3</sup> -CO] <sub>s</sub> -                      | (IIb), and    |
| -OC-R <sup>4</sup> -CO-                                      | (III)         |

in which units:

- ☐ X corresponds to the end function of said unit after reaction with an amine or carboxylic acid function;
- ☐ R<sup>1</sup>, which may be identical or different, [represent] represents a linear or branched alkyl radical comprising 2 to 4 carbon atoms;
- ☐ OA, which may be identical or different, [represent] represents an oxyethylenated, oxypropylenated or oxybutylenated radical, or mixtures thereof;
- ☐ R<sup>2</sup>, R<sup>2</sup>, R<sup>3</sup> and R<sup>4</sup>, which may be identical or different, represent:
  - a linear or branched alkyl radical comprising 2 to 18 carbon atoms,
  - an aryl radical comprising one or more optionally substituted aromatic nuclei,
- ☐ m is equal to 0 or 1,
- ☐ n is an integer between 4 and 800,
- ☐ r is such that the number of amide bonds is between 1 and 15 per unit (I),

**Attachment to Preliminary Amendment dated September 19, 2001**

**Marked-up Claims 1-23**

- ☐ s is such that the number of amide bonds is between 1 and 15 per unit (I),
- ☐ the molar ratio of the number of units (III) to the number of units (I) is between 0.5/1 and 1.4/1;
- ☐ the various units are linked together at least by means of amide bonds; and
- ☐ the number-average molar mass of the copolymer is less than 100 000 g/mol.

2. (Amended) The [use] gelling agent as claimed in [the preceding] claim 1, [characterized in that] wherein the units (I) of the copolymer comprise a sequence of oxyethylenated, oxypropylenated or oxyethylenated/oxypropylenated radicals, the sum of the units being equal to n.

3. (Amended) The [use] gelling agent as claimed in [either of the preceding claims] claim 1, [characterized in that] wherein the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, represent linear or branched radicals comprising 2 to 12 carbon atoms[, and preferably methylene radicals, optionally bearing one or more methyl radicals].

4. (Amended) The [use] gelling agent as claimed in [the preceding] claim 1, [characterized in that] wherein the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, are [chosen] selected from the group consisting of:

- ☐ ethyl, 1-methylethyl, propyl, 1-methylpropyl, butyl, hexyl, heptyl, octyl, decyl, undecyl and lauryl radicals.

**Attachment to Preliminary Amendment dated September 19, 2001**

**Marked-up Claims 1-23**

5. (Amended) The [use] gelling agent as claimed in [any one of the preceding claims] claim 1, [characterized in that] wherein the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, represent aryl radicals comprising one or more optionally substituted aromatic nuclei.

6. (Amended) The [use] gelling agent as claimed in [the preceding] claim 1, [characterized in that] wherein the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, comprise:

- ★ an aromatic nucleus, the reactive functions (amines or carboxylic acids) being in an ortho, meta or para position,
- ★ two aromatic nuclei, linked via inert groups, or peri-fused, [for instance divalent naphthyl radicals,] the reactive functions (amines or carboxylic acids) being on the carbon atoms 1 and 2, 1 and 4, 1 and 5, 1 and 6, 1 and 7 or 2 and 7.

7. (Amended) The [use] gelling agent as claimed in [any one of claims 1 to 4] claim 1, [characterized in that] wherein the units (IIa) or (IIb) are [chosen] selected from the group consisting of polyamide 6, polyamide 10, polyamide 11, polyamide 12, polyamide 6, 6 or a random copolymer of at least two such polyamides, in all proportions[, preferably 50/50].

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**Marked-up Claims 1-23**

8. (Amended) The [use] gelling agent as claimed in [any one of the preceding claims] claim 1, [characterized in that] wherein r and s, which may be identical or different, are such that the number of amide bonds is between 5 and 10 per unit (I).

9. (Amended) The [use] gelling agent as claimed in [any one of the preceding claims] claim 1, [characterized in that] wherein the number-average molecular mass of the copolymer is between 10 000 and 50 000 g/mol.

10. (Amended) The [use] gelling agent as claimed in [any one of the preceding claims] claim 1, [characterized in that] wherein the relatively nonpolar compound is in the Hansen solubility space, and has the following parameters:

- $\delta P$  of Keesom interactions of less than or equal to  $16.5 \text{ (J/cm}^3\text{)}^{1/2}$
- $\delta H$  of hydrogen bonds of less than or equal to  $10.5 \text{ (J/cm}^3\text{)}^{1/2}$
- $\delta D$  of London interactions of greater than or equal to  $15 \text{ (J/cm}^3\text{)}^{1/2}$ .

11. The [use] gelling agent as claimed in [any one of the preceding claims] claim 1, [characterized in that] wherein the nonionic surfactant is [chosen] selected from the group consisting of:

- polyoxyalkylenated (polyethoxyethylenated, polyoxypropylenated or polyoxybutylenated) alkylphenols in which the alkyl substituent is  $C_6-C_{12}$  and containing from 5 to 25 oxyalkylene units;

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**Marked-up Claims 1-23**

- polyoxyalkylenated C<sub>8</sub>-C<sub>22</sub> aliphatic alcohols containing from 1 to 25 oxyalkylene (oxyethylene or oxypropylene) units;
- products resulting from the condensation of ethylene oxide and/or propylene oxide with propylene glycol or ethylene glycol;
- ethoxylated and/or propoxylated C<sub>8</sub>-C<sub>18</sub> fatty acids containing from 5 to 25 ethoxylated and/or propoxylated units;
- alkoxyated amido amines containing from 1 to 50 oxyalkylenated units;
- alkoxyated terpenic hydrocarbons [such as ethoxylated and/or propoxylated  $\alpha$ - or  $\beta$ -pinenes], containing from 1 to 30 oxyethylene and/or oxypropylene units;
- alkylpolyglycosides which may be obtained by condensation of glucose with primary fatty alcohols containing a C<sub>4</sub>-C<sub>20</sub> alkyl group and also an average number of glucose units of about from 0.5 to 3 per mole of alkylpolyglycoside.

12. (Amended) The [use] gelling agent as claimed in [any one of the preceding claims] claim 1, [characterized in that] wherein the amount of copolymer relative to the relatively nonpolar compound or the nonionic surfactant is between 0.1 % and 15 % by weight [and preferably between 0.5 % and 10 % by weight].

**Attachment to Preliminary Amendment dated September 19, 2001**

**Marked-up Claims 1-23**

13. (Amended) [The use as claimed in any one of the preceding claims, characterized in that, in case of] A method for gelation of a relatively nonpolar compound, comprising using the gelling agent as claimed in claim 1, wherein the copolymer is combined with a nonionic or anionic surfactant.

14. (Amended) The [use] method as claimed in [the preceding] claim 13, [characterized in that] wherein the amount of nonionic or anionic surfactant relative to the relatively nonpolar compound is between 5% and 20% by weight.

15. (Amended) The [use] gelling agent as claimed in [any one of the preceding claims] claim 1, [characterized in that] wherein the copolymer is combined with a filler of lamellar structure.

16. (Amended) The [use] gelling agent as claimed in [the preceding] claim 15, [characterized in that] wherein the amount of filler represents up to 20% by weight of the copolymer.

17. (Amended) The [use] gelling agent as claimed in [either of claims 15 and 16] claim 15, [characterized in that] wherein the filler is introduced during the preparation of the copolymer and/or during the use of said copolymer.



**Attachment to Preliminary Amendment dated September 19, 2001**

**Marked-up Claims 1-23**

18. (Amended) [The use as claimed in any one of claims 1 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the] A composition of formulations intended for cleaning metals comprising an effective amount of the gelling agent according to claim 1.

19. (Amended) [The use as claimed in any one of claims 1 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the composition of] A detergent [formulations which may be] formulation used in the industrial field comprising an effective amount of the gelling agent according to claim 1.

20. (Amended) [The use as claimed in any one of claims 1 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the composition formulations] A formulation intended for stripping paints and varnishes comprising an effective amount of the gelling agent according to claim 1.

21. (Amended) [The use as claimed in any one of claims 15 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the composition of formulations] A method intended for cleaning or stripping vertical surfaces comprising using the gelling agent according to claim 15.

**Attachment to Preliminary Amendment dated September 19, 2001**

**Marked-up Claims 1-23**

22. (Amended) [The use as claimed in any one of claims 1 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the composition of formulations intended] A method for treating plants comprising using an effective amount of the gelling agent according to claim 1.

23. (Amended) [The use as claimed in any one of claims 1 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the] A composition [of formulations used in the field of] for ink printing [inks] comprising an effective amount of the gelling agent according to claim 1.

USE OF COPOLYMERS DERIVED FROM POLYAMIDES AS GELLING  
AGENTS FOR RELATIVELY NONPOLAR COMPOUNDS

The present invention relates to the use of copolymers comprising at least polyoxyalkylenated  
5 polyamide functions, as gelling agents for relatively nonpolar compounds or for surfactants, which are preferably nonionic.

Although there are many possibilities for preparing aqueous gels, gels made of relatively  
10 nonpolar compounds or of surfactants have not been particularly developed, although many fields may be interested by the production of such gels. This is the case especially for the cosmetics field (for example suncreams); the field of industrial cleaning/stripping  
15 (treatment of vertical surfaces); the field of plant protection (maintenance in suspension of hydrolysis-sensitive active materials, for example); the field of printing inks; the field of detergency; the field of public works and construction; the papermaking field  
20 (for example the formulation of antifoams for manufacturing paper pulp); the explosives field (for example the gelation of ergols or propergols for military and civil applications) or the field of hydraulic fluids.

25 The aim of the present invention is to propose a simple and efficient means for gelling relatively nonpolar media or surfactant media.

Thus, one subject of the invention is the use as a gelling agent, of a relatively nonpolar compound or a nonionic surfactant, of at least one block copolymer comprising the following units:

- $$\begin{array}{ll}
5 & -X-(R^1)_m-(OA)_n-X- & \text{(I), and} \\
& -[NH-R^2-NHCO-R'^2-CO]_x- & \text{(IIa), and/or} \\
& -[NH-R^3-CO]_s- & \text{(IIb), and} \\
& -OC-R^4-CO- & \text{(III)}
\end{array}$$

in which units:

- 10    □ X corresponds to the end function of said unit after  
reaction with an amine or carboxylic acid function;  
□ R<sup>1</sup>, which may be identical or different, represent a  
linear or branched alkyl radical comprising 2 to 4  
carbon atoms;
- 15    □ OA, which may be identical or different, represent an  
oxyethylenated, oxypropylenated or oxybutylenated  
radical, or mixtures thereof;  
□ R<sup>2</sup>, R'<sup>2</sup>, R<sup>3</sup> and R<sup>4</sup>, which may be identical or  
different, represent:
- 20    • a linear or branched alkyl radical comprising 2 to  
18 carbon atoms,  
• an aryl radical comprising one or more optionally  
substituted aromatic nuclei,
- m is equal to 0 or 1,
- 25    □ n is an integer between 4 and 800,  
□ r is such that the number of amide bonds is between 1  
and 15 per unit (I),  
□ s is such that the number of amide bonds is between 1

and 15 per unit (I),

- the molar ratio of the number of units (III) to the number of units (I) is between 0.5/1 and 1.4/1;
- the various units are linked together at least by means of amide bonds;
- the number-average molar mass of the copolymer is less than 100 000 g/mol.

However, other characteristics will emerge more clearly on reading the description and the examples which follow.

Before describing the copolymer used in the invention in more precise detail, the nature of the compound which may be gelled according to the invention will be given.

Thus, the expression "relatively nonpolar compound" means any constituent which is liquid at the temperature for preparing and/or using the formulation of which it forms one of the constituent elements.

Moreover, this liquid, which is in the Hansen solubility space (Handbook of solubility parameters and other cohesion parameters - Allan F.M. Barton, CRC Press Inc., 1983-), more particularly has the following parameters:

.  $\delta P$  of Keesom interactions of less than or equal to

25  $16.5 \text{ (J/cm}^3\text{)}^{1/2}$

.  $\delta H$  of hydrogen bonds of less than or equal to 10.5  $\text{(J/cm}^3\text{)}^{1/2}$

.  $\delta D$  of London interactions of greater than or equal to

15  $(\text{J}/\text{cm}^3)^{1/2}$ .

More particularly, the coefficient  $\delta P$  is between 2 and  $16.5 (\text{J}/\text{cm}^3)^{1/2}$ .

Preferably, the coefficient  $\delta P$  is between 2 and  $10.5 (\text{J}/\text{cm}^3)^{1/2}$ .

It should be noted that the coefficient  $\delta D$  is usually between 15 and 23.

As non-limiting examples of relatively nonpolar compounds, mention may be made of:

- 10 . triglycerides of saturated or unsaturated fatty acids containing at least 12 carbon atoms and preferably from 14 to 20 carbon atoms; these may be synthetic or, preferably, natural triglycerides, for instance plant oils such as rape seed oil, soybean oil,
- 15 groundnut oil, butter oil, cottonseed oil, flax oil, coconut oil, olive oil, palm oil, grapeseed oil, fish oil, castor oil or copra oil;
- . methyl esters of saturated or unsaturated fatty acids containing at least 12 carbon atoms and preferably
- 20 from 14 to 20 carbon atoms, for instance methyl oleate;
- . aliphatic or cyclic  $\text{C}_1\text{-C}_8$  alkyl esters of saturated monocarboxylic acids, for instance methyl acetate, ethyl acetate or cyclohexyl acetate;
- 25 . terpenic compounds (D-limonene, L-limonene, etc.);
- .  $\text{C}_1\text{-C}_4$  alkyl diesters of at least one  $\text{C}_4\text{-C}_6$  aliphatic diacid. Mixtures of diacid esters which are esters derived essentially from adipic acid, glutaric acid

and succinic acid are more particularly used, the alkyl groups of the ester portion being chosen especially from methyl and ethyl groups, but may also be propyl, isopropyl, butyl, n-butyl and isobutyl;

- 5 . anisole;
- . n-methylpyrrolidone;
- . dimethyl sulfoxide;
- . ketones, for instance cyclopentanone or methyl isobutyl ketone;
- 10 . polyalkylene glycols, for instance polyethylene glycol 400 or polypropylene glycol 400.

The present invention may be used for gelling a relatively nonpolar compound or a mixture of several such compounds.

- 15 Surfactants, and more particularly nonionic surfactants, may similarly be gelled according to the process of the present invention.

More particularly, the surfactant is in the form of a liquid at the temperature for preparing and/or using the formulation of which it forms one of the constituent elements.

- In addition, the surfactant is preferably free of polar solvents, but, if such a solvent is present, the solvent content in this surfactant remains such that it does not prevent the subsequent gelation of said surfactant.
- 25

By way of nonionic surfactant which is most particularly suitable for the invention, mention may be

made, without, however, intending to be limited thereto, of:

- . polyoxyalkylenated (polyethoxyethylenated, polyoxypropylenated or polyoxybutylenated)
- 5    alkylphenols in which the alkyl substituent is  $C_6-C_{12}$  and containing from 5 to 25 oxyalkylene units; examples which may be mentioned are Triton X-45, X-114, X-100 or X-102 sold by Rohm & Haas Co. and Igepal NP2 to NP17 from Rhodia Chimie;
- 10    . polyoxyalkylenated  $C_8-C_{22}$  aliphatic alcohols containing from 1 to 25 oxyalkylene (oxyethylene or oxypropylene) units; examples which may be mentioned are the products Tergitol 15-S-9 and 24-L-6 NMW sold by Union Carbide Corp., Neodol 45-9, 23-65, 45-7 and
- 15    45-4 sold by Shell Chemical Co., Kyrol EOB sold by Procter & Gamble Co., Synperonic A3 to A9 from ICI and Rhodasurf IT, DB and B from Rhodia Chimie;
- . products resulting from the condensation of ethylene oxide and/or propylene oxide with propylene glycol or
- 20    ethylene glycol, with a weight-average molecular mass of about from 2 000 to 10 000, such as the Pluronic products sold by BASF;
- . alkoxyated amido amines containing from 1 to 50, preferably from 1 to 25 and most particularly from 2
- 25    to 20 oxyalkylene (preferably oxyethylene) units;
- . alkoxyated terpenic hydrocarbons such as ethoxyated and/or propoxyated  $\alpha$ - or  $\beta$ -pinenes, containing from 1 to 30 oxyethylene and/or oxypropylene units; in

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particular such as those described in international application WO 96/01245, to which reference may be made in particular;

. alkylpolyglycosides which may be obtained by

- 5      condensation (for example by acid catalysis) of  
glucose with primary fatty alcohols (US-A-3 598 865;  
US 4 565 647; EP 132 043; EP 132 046, etc.)  
containing a C<sub>4</sub>-C<sub>20</sub> and preferably C<sub>8</sub>-C<sub>18</sub> alkyl group  
and also an average number of glucose units of about  
10    from 0.5 to 3 and preferably of about from 1.1 to 1.8  
per mole of alkylpolyglycoside (APG); mention may be  
made in particular of those containing a C<sub>8</sub>-C<sub>14</sub> alkyl  
group and on average 1.4 glucose units per mole; a  
C<sub>12</sub>-C<sub>14</sub> alkyl group and on average 1.4 glucose units  
15    per mole; a C<sub>8</sub>-C<sub>14</sub> alkyl group and on average 1.5  
glucose units per mole; a C<sub>8</sub>-C<sub>10</sub> alkyl group and on  
average 1.6 glucose units per mole; which are sold,  
respectively, under the names GlucoPON 600 EC®, 600  
CSUP®, 650 EC® and 225 CSUP® by Henkel.

- 20            The present invention makes it possible to  
gel one of the surfactants mentioned above, and also  
mixtures thereof.

The copolymer will now be described.

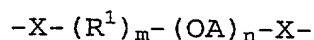
- As mentioned above, the copolymer comprises  
25    units (I) to (III), which are more particularly  
randomly distributed.

The copolymer comprises amide bonds, ether  
bonds arising from the unit (I) and, depending on the

nature of the reactive functions of the unit (I), possibly ester and urethane bonds.

Moreover, the end functions of this copolymer are amines, more particularly primary amines,  
 5 carboxylic acids and optionally alcohols.

The copolymer comprises at least one unit (I) of the following formula:



in which formula:

- 10 ☐ X corresponds to the end function of said unit after reaction with an amine or carboxylic acid function;
- ☐  $R^1$ , which may be identical or different, represent a linear or branched alkyl radical comprising 2 to 4 carbon atoms;
- 15 ☐ OA, which may be identical or different, represent an oxyethylenated, oxypropylenated or oxybutylenated radical, or mixtures thereof;
- ☐ m is equal to 0 or 1;
- ☐ n is an integer between 4 and 800.

20 According to one more particular embodiment of the invention, the unit (I) of the copolymer comprises a sequence of oxyethylenated, oxypropylenated or oxybutylenated radicals, or a combination of two or three of these units, the sum of the units being equal  
 25 to n.

It should be noted that the copolymers forming the subject of the use according to the invention may, depending on their subsequent use, have

their structure adapted, especially via the nature and number of oxyalkylenated units. Thus, for example, the compatibility of the copolymer with a very sparingly polar compound may be increased by reducing the number of oxyethylenated radicals and/or by increasing the number of oxypropylenated or even oxybutylenated radicals.

Preferably, the number of units OA is between 4 and 300.

10 X corresponds to the end function of said unit after reaction with an amine or carboxylic acid function. To be more precise, X is obtained from the reaction of monomer(s) or of prepolymer(s), which are, for the copolymer forming the subject of the invention, 15 the source of the unit (I), with an amine or carboxylic acid function.

It should be noted that it is advantageous to use prepolymers corresponding to the unit (I) which are obtained extemporaneously, and which contain end 20 functions capable of reacting with amine or carboxylic acid functions.

Among these suitable end functions, mention may be made most particularly of alcohols, amines, carboxylic acids, epoxides and isocyanates.

25 Thus, when X is obtained from the reaction of an amine end function with a carboxylic acid, that is to say an amide function, X corresponds to the following function:  $\text{-NH-}$ . When X is obtained from the

reaction of an alcohol end function with a carboxylic acid, that is to say an ester function, X corresponds to the following function: -O-; and so on.

According to a first preferred embodiment of the invention, the unit (1) contains a group X resulting from the reaction of an amine function with a carboxylic acid, in which case m is 1.

According to this particular embodiment, when the radical OA is an oxyethylenated radical, the radical  $R^1$  may contain 2 to 4 carbon atoms. When the radical OA is an oxypropylenated or oxybutylenated radical, the radical  $R^1$  more particularly comprises 3 or 4 carbon atoms, respectively.

Such units (I) derive from polyoxyalkylenated diamines. These compounds are well known to those skilled in the art and are sold in particular under the brand name Jeffamine® (Texaco-Huntsmann).

According to a second preferred embodiment of the invention, the unit (I) contains a group X resulting from the reaction of an alcohol with a carboxylic acid, in which case m is preferably 0.

The units (I) corresponding to this variant are obtained from block or random polyalkylene glycols or polyalkylene glycol copolymers.

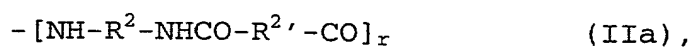
Compounds which may be used in the present invention are sold in particular under the brand name Pluronic PE® (BASF).

When the unit (I) contains a group X

resulting from the reaction of a epoxide or an isocyanate with an amine or a carboxylic acid, then m is preferably equal to 0.

The copolymer according to the invention may  
 5 comprise only one type of unit (I) or a combination of several such units, whether these units are differentiated by the nature of their units X and/or of their units OA.

The second types of units of the copolymers  
 10 used in the invention have the following formulae:



and/or



in which formulae:

15  $\square$   $\text{R}^2$ ,  $\text{R}'^2$ ,  $\text{R}^3$  and  $\text{R}^4$ , which may be identical or different, represent:

- a linear or branched alkyl radical comprising 2 to 18 carbon atoms,
- an aryl radical comprising one or more optionally  
 20 substituted aromatic nuclei,

$\square$  r is such that the number of amide bonds is between 1 and 15 per unit (I),

$\square$  s is such that the number of amide bonds is between 1 and 15 per unit (I).

25 More particularly, the radicals  $\text{R}^2$ ,  $\text{R}'^2$  and  $\text{R}^3$ , which may be identical or different, represent linear or branched radicals comprising 2 to 12 carbon atoms, and preferably methylene radicals, optionally

bearing one or more methyl radicals.

According to one preferred embodiment, said radicals, which may be identical or different, are chosen in particular from divalent ethyl,

5 1-methylethyl, propyl, 1-methylpropyl, butyl, hexyl, heptyl, octyl, decyl, undecyl and lauryl radicals.

Another possibility consists of radicals  $R^2$ ,  $R'^2$  and  $R^3$ , which may be identical or different, representing aryl radicals comprising one or more  
10 optionally substituted aromatic nuclei.

When the abovementioned radicals comprise only one aromatic nucleus, preferably containing 6 carbon atoms, the reactive functions (carboxylic acids and/or amines) are in an ortho, meta or para position.

15 It should be noted that when the above-mentioned radicals comprise several aromatic nuclei, preferably two aromatic nuclei, these nuclei may be peri-fused or linked via inert groups, such as simple valency bonds or an alkyl radical comprising 1 to 4  
20 carbon atoms.

Among the radicals comprising two aromatic nuclei, mention may be made most particularly of divalent naphthyl radicals attached to the reactive functions (carboxylic acids and/or amines) via the  
25 carbon atoms 1 and 2, 1 and 4, 1 and 5, 1 and 6, 1 and 7 or 2 and 7.

The values of  $r$  and  $s$  are more particularly between 5 and 10, per unit (I).

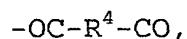
The units (IIa) are obtained by reacting a diamine with a diacid, and the units (IIb) are obtained by reacting amino acids or lactams.

Such units may be obtained in situ, during  
5 the preparation of the copolymer used in the invention, or alternatively may be prepared extemporaneously: these units are produced in accordance with a conventional polyamidation process, which will be described in detail hereinbelow.

10 Thus, according to one preferred variant of the invention, the units used as units (IIa) or (IIb) are polyamides, such as polyamide 6, polyamide 10, polyamide 11, polyamide 12 or polyamide 6,6, or a random copolymer of at least two such polyamides, in  
15 all proportions, preferably 50/50.

It should be noted that the use of units (IIa) and (IIb) which have just been described has the additional advantage of making it possible to obtain a copolymer whose melting point is not very high, thus  
20 facilitating the preparation of the gel, during which the polymer is molten.

As regards the units (III), their formula is as follows:



25 in which  $\text{R}^4$ , which may be identical or different, have the same meaning as that given for the radicals  $\text{R}^2$  to  $\text{R}^3$  mentioned above. Reference may thus be made thereto.

The units (III) are obtained from the

corresponding diacids.

The molar ratio of the number of units (III) to the sum of the number of units (I) is between 0.5/1 and 1.4/1. According to one preferred embodiment of the invention, the number of units (III) is such that it is close to the stoichiometry between the number of amine, carboxylic and optionally alcohol, isocyanate and epoxy functions.

The copolymers, or the mixture of copolymers, used in the invention also have the advantage of having a relatively low melting point, that is to say a melting point of less than about 190°C, more particularly between 70 and 190°C and preferably between 100 and 190°C.

The copolymers which have just been described may be prepared by reacting dicarboxylic acids, polyoxyalkylenated compounds containing end functions capable of reacting with amine or carboxylic acid functions (in the form of monomers or prepolymers), amino acids or lactams, or alternatively polyamides.

The process is performed by placing the various reagents in contact, preferably in the absence of solvent.

This first step is generally performed without heating. However, when one or more of the reagents is in a solid form, the operation is performed at a temperature at least equal to the highest melting point.



The various reagents are used in amounts that are suitable for obtaining a copolymer as defined above.

Once the reagents have been placed in  
5 contact, the actual polymerization reaction is performed. To do this, the temperature is raised to a value generally of between 100°C and 300°C. During this step, the reaction water is evaporated off.

The pressure conditions during the  
10 polymerization reaction may be readily determined by a person skilled in the art.

A catalyst may optionally be used to promote the polyamidation reaction, such as, for example, hypophosphorous acid, phenylphosphinic acid,  
15 phenylphosphonic acid or tris(nonylphenyl) phosphite.

A catalyst which promotes polyesterification may also be used, if necessary. Among the catalysts which may be used, mention may be made of titanium-based and zirconium-based catalysts, inter alia, such  
20 as, for example, titanium tetrabutyl orthotitanate and tetrabutyl orthozirconate.

The catalyst may represent from 0% to 1% by weight relative to the total weight of the various monomers, more particularly up to 0.5% and preferably  
25 between a few tens and a few hundreds of parts per million.

As has been mentioned previously, the copolymers used in the invention have a number-average

molar mass of less than or equal to 100 000 g/mol and more particularly between 10 000 and 50 000 g/mol.

It should be noted that the number-average molar masses of the copolymers were evaluated by steric exclusion chromatography, using polystyrene as standard.

The copolymers are more particularly used in a proportion of from 0.1% to 15% by weight relative to the relatively nonpolar compound or the surfactant.

10 Advantageously, the copolymer content is between 0.5 and 10% by weight relative to the same reference.

In the case of a gelation of a relatively nonpolar compound, it should be noted that the copolymer used in the present invention may be combined

15 with at least one nonionic or anionic surfactant, in order to make said relatively nonpolar compound self-emulsifying.

In such a case, the surfactant is preferably free of polar solvent or, in any case, the solvent

20 content in this surfactant, if it is present, should be such that it does not prevent the subsequent gelation of the copolymer/surfactant/relatively nonpolar compound mixture.

Among the nonionic surfactants which are

25 suitable, mention may be made of:

- . polyoxyalkylenated (polyethoxyethylenated, polyoxypropylenated or polyoxybutylenated)
- alkylphenols in which the alkyl substituent is C<sub>6</sub>-C<sub>12</sub>

and containing from 5 to 25 oxyalkylene units;  
examples which may be mentioned are Triton X-45,  
X-114, X-100 or X-102 sold by Rohm & Haas Co. and  
Igepal NP2 to NP17 from Rhodia Chimie;

- 5 . polyoxyalkylenated  $C_8-C_{22}$  aliphatic alcohols  
containing from 1 to 25 oxyalkylene (oxyethylene or  
oxypropylene) units; examples which may be mentioned  
are the products Tergitol 15-S-9 and 24-L-6 NMW sold  
by Union Carbide Corp., Neodol 45-9, 23-65, 45-7 and  
10 45-4 sold by Shell Chemical Co., Kyro EOB sold by  
Procter & Gamble Co., Synperonic A3 to A9 from ICI  
and Rhodasurf IT, DB and B from Rhodia Chimie;
- . ethoxylated or ethoxy-propoxylated triglycerides of  
plant or animal origin, such as, for example, lard,  
15 tallow, groundnut oil, butter oil, cottonseed oil,  
flax oil, olive oil, palm oil, grape seed oil, fish  
oil, soybean oil, castor oil, rape seed oil, copra  
oil or coconut oil;
- . products resulting from the condensation of ethylene  
20 oxide and/or propylene oxide with propylene glycol or  
ethylene glycol, with a weight-average molecular mass  
of about from 2 000 to 10 000, such as the Pluronic  
products (BASF);
- . products resulting from the condensation of ethylene  
25 oxide and/or propylene oxide with ethylenediamine,  
such as the Tetronic products (BASF);
- . ethoxylated and/or propoxylated  $C_8-C_{18}$  fatty acids  
containing from 5 to 25 ethoxylated and/or

- propoxylated units;
- . C<sub>8</sub>-C<sub>20</sub> fatty acid amides containing from 5 to 30 ethoxylated units;
  - . ethoxylated amines containing from 5 to 30
  - 5 ethoxylated units;
  - . alkoxyated amido amines containing from 1 to 50, preferably from 1 to 25 and most particularly from 2 to 20 oxyalkylene units (preferably oxyethylene units);
  - 10 . amine oxides such as (C<sub>10</sub>-C<sub>18</sub>)alkyl dimethylamine oxides and (C<sub>8</sub>-C<sub>22</sub>)alkoxyethyldihydroxyethylamine oxides;
  - . alkoxyated terpenic hydrocarbons such as ethoxylated and/or propoxylated  $\alpha$ - or  $\beta$ -pinenes, containing from
  - 15 1 to 30 oxyethylene and/or oxypropylene units; for instance those described in patent WO 96/01245;
  - . alkylpolyglycosides which may be obtained by condensation (for example by acid catalysis) of glucose with primary fatty alcohols (US-A-3 598 865;
  - 20 US 4 565 647; EP 132 043; EP 132 046, etc.) containing a C<sub>4</sub>-C<sub>20</sub> and preferably C<sub>8</sub>-C<sub>18</sub> alkyl group and also an average number of glucose units of about from 0.5 to 3 and preferably of about from 1.1 to 1.8 per mole of alkylpolyglycoside (APG); mention may be
  - 25 made in particular of those containing a C<sub>8</sub>-C<sub>14</sub> alkyl group and on average 1.4 glucose units per mole; a C<sub>12</sub>-C<sub>14</sub> alkyl group and on average 1.4 glucose units per mole; a C<sub>8</sub>-C<sub>14</sub> alkyl group and on average 1.5

glucose units per mole; a C<sub>8</sub>-C<sub>10</sub> alkyl group and on average 1.6 glucose units per mole; which are sold, respectively, under the names GlucoPON 600 EC®, 600 CSUP®, 650 EC® and 225 CSUP® by Henkel.

5           As regards the anionic surfactants, use is made more particularly of surfactants which are in the form of liquid or in a form which is soluble in the relatively nonpolar compound. Examples which may be mentioned are:

- 10 . alkyl ester sulfonates of formula R-CH(SO<sub>3</sub>M)-COOR', in which R represents a C<sub>8</sub>-C<sub>20</sub> and preferably C<sub>10</sub>-C<sub>16</sub> alkyl radical, R' represents a C<sub>1</sub>-C<sub>6</sub> and preferably C<sub>1</sub>-C<sub>3</sub> alkyl radical and M represents a cation chosen from an alkali metal or alkaline-earth metal (sodium, potassium, lithium or calcium), a substituted or  
15 unsubstituted ammonium residue (methyl-, dimethyl-, trimethyl- or tetramethylammonium, dimethyl-piperidinium, etc.) or an alkanolamine derivative (monoethanolamine, diethanolamine, triethanolamine, etc.);  
20 etc.);
- . alkyl sulfates of formula ROSO<sub>3</sub>M, in which R represents a C<sub>5</sub>-C<sub>24</sub> and preferably C<sub>10</sub>-C<sub>18</sub> alkyl or hydroxyalkyl radical, M representing a hydrogen atom or a cation as defined above, and also the  
25 ethoxylated (EO) and/or propoxylated (PO) derivatives thereof, containing on average from 0.5 to 30 and preferably from 0.5 to 10 EO and/or PO units;

- . alkylamide sulfates of formula  $RCONHR'OSO_3M$  in which R represents a  $C_2-C_{22}$  and preferably  $C_6-C_{20}$  alkyl radical,  $R'$  represents a  $C_2-C_3$  alkyl radical, M representing a hydrogen atom or a cation of the same definition as above, and also the ethoxylated (EO) and/or propoxylated (PO) derivatives thereof, containing on average from 0.5 to 60 EO and/or PO units;
- . alkoxyated (ethoxylated and/or propoxylated)
- 10 phosphate esters derived from aliphatic alcohols, from alkylphenols or from alkylarylphenols;
- . saturated or unsaturated  $C_8-C_{24}$  and preferably  $C_{14}-C_{20}$  fatty acid salts,  $C_9-C_{20}$  alkylbenzenesulfonates, primary or secondary  $C_8-C_{22}$  alkylsulfonates,
- 15 alkylglyceryl sulfonates, the sulfonated polycarboxylic acids described in GB-A-1 082 179, paraffin sulfonates, N-acyl N-alkyltaurates, alkyl phosphates, isethionates, alkylsuccinamates, alkylsulfosuccinates, sulfosuccinate monoesters or
- 20 diesters, N-acyl sarcosinates, alkylglycoside sulfates and polyethoxycarboxylates;
- . sophorolipids, such as those in acid or lactone form, derived from 17-hydroxyoctadecenoic acid;
- . the associated cation has the same definition as that
- 25 given for M, or is a hydrogen atom.

When a surfactant is present, the amount used is more particularly between 5% and 20% by weight relative to the relatively nonpolar compound.

The advantage of introducing a surfactant into the copolymer/relatively nonpolar compound mixture is that it makes the resulting gel self-emulsifying.

The copolymer according to the invention may  
5 similarly be used with a filler such as, for example, a filler of lamellar structure. By way of illustration, smectites (montmorillonite, beidellite, nontronite, hectorite, saponite, etc.) may be used.

The filler content varies within a wide  
10 range. Advantageously, and when it is present, its content may represent up to 20% by weight of the copolymer.

This filler may be used in several ways.

The first consists in using it during the  
15 preparation of the copolymer. This possibility has the surprising advantage of reducing the melting point of the copolymer thus obtained. This may prove to be advantageous when using the copolymer to prepare the gel.

20 The second consists in adding the filler when using the copolymer. Such an operation makes it possible to increase the hardness of the resulting gel.

Needless to say, it would not constitute a departure from the context of the present invention to  
25 combine these two variants.

One of the fields of application of the mixture comprising the copolymer and the relatively nonpolar compound or the surfactant relates to

formulations intended for cleaning or stripping paints and varnishes, on various surfaces, for example such as metals.

The advantage of this mixture according to  
5 the invention is that it makes it possible to obtain a highly viscous composition which makes the cleaning/stripping operation more efficient by means of better "attachment" of the active composition to the support to be treated.

10 The mixtures according to the invention may be used, for example, for cleaning/stripping vertical surfaces.

Another field of application of the mixture according to the invention relates to that of  
15 industrial detergency, for which it makes it possible to provide a means for gelling solutions comprising, for example, compounds of the type such as limonenes.

Similarly, it makes it possible to gel liquid additives for solid detergent formulations.

20 The mixture according to the invention may also be used in the plant protection field.

It may be advantageous to have available gelled mixtures comprising one or more active materials that are liquid, dissolved or held in suspension, in a  
25 suitable solvent, from the moment that these mixtures show solubility parameters within the ranges indicated above.

Specifically, such mixtures are stable up to



temperatures of at least about 50°C.

The mixture according to the invention may also find an application in the field of printing inks, with the gelation of mineral or plant oils, inter alia.

5           The mixtures according to the invention are obtained by placing the copolymer in contact with the relatively nonpolar compound or the surfactant and, where appropriate, the abovementioned additives.

          This placing in contact preferably takes  
10 place with heating over a period which is sufficient to dissolve or disperse the (co)polymer. For example, a temperature of at least 90°C is suitable for carrying out the invention. Advantageously, the dissolution or dispersion may be carried out at a temperature at which  
15 the copolymer is in molten form.

          In the case of the gelation of relatively nonpolar compounds, it may be that the melting point of the copolymers described previously is higher than the flash point of said relatively nonpolar compounds.  
20 Consequently, the operation of placing in contact may take place preferably under a stream of nitrogen.

          The time for which the compounds are placed in contact is more particularly such that the copolymer is dissolved.

25           Once dissolution has been achieved, the mixture is cooled without stirring to facilitate the formation of the gel.

Concrete but nonlimiting examples of the

invention will now be given.

### EXAMPLE 1

#### 1) Preparation of the copolymer

The following reagents are placed in a glass  
5 reactor:

Compound	% by weight	Number of mols
Jeffamine ED 2003 (*)	76.1	1
Adipic acid	4.9	1
Aminocaproic acid	9.5	2.19
Aminoundecanoic acid	9.5	1.43
H <sub>3</sub> PO <sub>2</sub> catalyst	500 ppm	

(\*) Jeffamine ED 2003 (Huntsmann); Mn = 2276 g/mol; molar  
ratio of ethylene oxide (EO)/propylene oxide  
(PO) = 37.5/5.5;

10

The reactor is then purged (vacuum/nitrogen)  
and the temperature is raised to 230°C, under nitrogen.  
After maintenance at this temperature for one hour, a  
vacuum of 1 mbar is gradually applied, over half an  
15 hour, at 230°C. These conditions are maintained for  
10 minutes.

A block copolymer comprising on average 4  
amide bonds per polyamide block (determined by NMR) is  
obtained. In addition, the number-average molar mass is  
20 about 46 400 g/mol (determined by steric exclusion  
chromatography; polystyrene standard).

#### 2) Preparation of a gel

The copolymer obtained above is dissolved

while hot (above the melting of the copolymer), under a stream of nitrogen, into Rhodiasolv RPDE, sold by Rhodia Chimie (mixture of short-chain adipic acid, glutaric acid and succinic acid esters). The copolymer is present at 1% by weight relative to the Rhodiasolv RPDE.

Once the dissolution has been achieved, the mixture is cooled without stirring to facilitate the formation of the gel.

The gel is stable up to 50°C.

## EXAMPLE 2

### 1) Preparation of the copolymer

The process is performed as in example 1, except that the composition used is as follows:

Compound	% by weight	Number of mols
Jeffamine D 2000(*)	66.6	1
Adipic acid	4.85	1
Aminocaproic acid	28.5	6.59
H <sub>3</sub> PO <sub>2</sub> catalyst	500 ppm	

(\*) Jeffamine D 2000 (Texaco); Mn = 2202 g/mol; polypropylene oxide.

A block copolymer comprising on average 5 amide bonds per polyamide block (determined by NMR) is obtained. In addition, the number-average molar mass is about 17 200 g/mol (determined by steric exclusion chromatography; polystyrene standard).

### 2) Preparation of a gel

The copolymer obtained above is dissolved

while hot, under a stream of nitrogen, into Phylorob 926-67 methyl oleate sold by Novance. The copolymer is present at 1% by weight relative to the methyl oleate.

Once the dissolution is achieved, the mixture  
5 is cooled without stirring.

The gel is stable up to 50°C.

### EXAMPLE 3

#### 1) Preparation of the copolymer with a mineral filler

The process is performed as in example 1,  
10 except that the composition used is as follows:

Compound	% by weight	Number of mols
Jeffamine D 2000 (*)	66.6	1
Adipic acid	4.85	1
Aminocaproic acid	14.3	3.3
Aminoundecanoic acid	14.3	2.15
H <sub>3</sub> PO <sub>2</sub> catalyst	500 ppm	
Montmorillonite (**)	X	

(\*) Jeffamine D 2000 (Texaco); Mn = 2002 g/mol;  
polypropylene oxide

(\*\*) Montmorillonite SCPX 1789 from Laporte, untreated.

15 X represents 0%, 5% or 20% by weight relative to the weight of the copolymer.

A block copolymer comprising on average 5.5 amide bonds per polyamide block (determined by NMR) is obtained.

2) Measuring the melting points

Montmorillonite content	Melting point
0%	127°C
5%	116°C
20%	111°C

**EXAMPLE 4**

5           The copolymer obtained in example 1 is dissolved while hot (above the melting point of the copolymer), under a stream of nitrogen, in PPG 400 (polypropylene glycol with a weight-average molar mass of 400 - sold by Dow). The copolymer is present at 5%  
10 by weight relative to the PPG 400.

Once the dissolution is achieved, the mixture is cooled without stirring to facilitate formation of the gel.

The gel is stable up to 50°C.

15           A similar result is obtained using PEG 400 (polyethylene glycol with a weight-average molar mass of 400 - sold by Dow) with 2% of copolymer.

**EXAMPLE 5**

20           The copolymer obtained in example 1 is dissolved while hot in Synperonic A7 (C<sub>12</sub>-C<sub>14</sub> fatty alcohol comprising 7 oxyethylenated units - sold by ICI). The copolymer is present at 2% by weight relative to the Synperonic A7.

A gel is obtained once dissolution has been

achieved, and the mixture has been cooled without stirring to facilitate formation of the gel.

**EXAMPLE 6**

The copolymer obtained in example 1 is  
5 dissolved while hot, in nonylphenol 10 EO (Igepal N10 - Rhodia Chimie). The copolymer is present at 5% by weight relative to the nonylphenol.

A gel is obtained once dissolution has been achieved and the mixture has been cooled without  
10 stirring to facilitate formation of the gel.

**EXAMPLE 7**

The copolymer obtained in example 1 is dispersed while hot in an alkylpolyglycoside comprising a C<sub>12</sub>-C<sub>16</sub> alkyl unit (Plantaren 1200). The copolymer is  
15 present at 5% by weight relative to the Plantaren.

A gel is obtained once the dispersion has been achieved and the mixture has cooled without stirring to facilitate formation of the gel.

**EXAMPLE 8**

20 The copolymer obtained in example 1 is dissolved while hot in a nopol containing 3 propylene oxide (PO) units and 6.5 ethylene oxide (EO) units. This product is obtained by carrying out the teaching of international application WO 96/01245.

25 A gel is obtained once dissolution has been achieved and the mixture has cooled without stirring to facilitate formation of the gel.

10    □ X corresponds to the end function of said unit after  
reaction with an amine or carboxylic acid function;  
□ R<sup>1</sup>, which may be identical or different, represent a  
linear or branched alkyl radical comprising 2 to 4  
carbon atoms;

15    □ OA, which may be identical or different, represent an  
oxyethylenated, oxypropylenated or oxybutylenated  
radical, or mixtures thereof;  
□ R<sup>2</sup>, R'<sup>2</sup>, R<sup>3</sup> and R<sup>4</sup>, which may be identical or  
different, represent:

20    • a linear or branched alkyl radical comprising 2 to  
18 carbon atoms,  
• an aryl radical comprising one or more optionally  
substituted aromatic nuclei,

□ m is equal to 0 or 1,

25    □ n is an integer between 4 and 800,  
□ r is such that the number of amide bonds is between 1  
and 15 per unit (I),

- s is such that the number of amide bonds is between 1 and 15 per unit (I),
- the molar ratio of the number of units (III) to the number of units (I) is between 0.5/1 and 1.4/1;
- 5 □ the various units are linked together at least by means of amide bonds;
- the number-average molar mass of the copolymer is less than 100 000 g/mol.

2. The use as claimed in the preceding  
10 claim, characterized in that the units (I) of the copolymer comprise a sequence of oxyethylenated, oxypropylenated or oxyethylenated/oxypropylenated radicals, the sum of the units being equal to n.

3. The use as claimed in either of the  
15 preceding claims, characterized in that the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, represent linear or branched radicals comprising 2 to 12 carbon atoms, and preferably methylene radicals, optionally bearing one or more methyl radicals.

20 4. The use as claimed in the preceding claim, characterized in that the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, are chosen from:

- ethyl, 1-methylethyl, propyl, 1-methylpropyl,
- 25 butyl, hexyl, heptyl, octyl, decyl, undecyl and lauryl radicals.

5. The use as claimed in any one of the preceding claims, characterized in that the radicals  $R^2$ ,



$R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, represent aryl radicals comprising one or more optionally substituted aromatic nuclei.

6. The use as claimed in the preceding  
5 claim, characterized in that the radicals  $R^2$ ,  $R'^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, comprise:
- \* an aromatic nucleus, the reactive functions (amines or carboxylic acids) being in an ortho, meta or para position,
  - 10 \* two aromatic nuclei, linked via inert groups, or peri-fused, for instance divalent naphthyl radicals, the reactive functions (amines or carboxylic acids) being on the carbon atoms 1 and 2, 1 and 4, 1 and 5, 1 and 6, 1 and 7 or 2 and 7.

- 15 7. The use as claimed in any one of claims 1 to 4, characterized in that the units (IIa) or (IIb) are chosen from polyamide 6, polyamide 10, polyamide 11, polyamide 12, polyamide 6,6 or a random copolymer of at least two such polyamides, in all  
20 proportions, preferably 50/50.

8. The use as claimed in any one of the preceding claims, characterized in that  $r$  and  $s$ , which may be identical or different, are such that the number of amide bonds is between 5 and 10 per unit (I).

- 25 9. The use as claimed in any one of the preceding claims, characterized in that the number-average molecular mass of the copolymer is between 10 000 and 50 000 g/mol.

10. The use as claimed in any one of the preceding claims, characterized in that the relatively nonpolar compound is in the Hansen solubility space, and has the following parameters:

- 5 .  $\delta P$  of Keesom interactions of less than or equal to  $16.5 \text{ (J/cm}^3\text{)}^{1/2}$
- .  $\delta H$  of hydrogen bonds of less than or equal to  $10.5 \text{ (J/cm}^3\text{)}^{1/2}$
- .  $\delta D$  of London interactions of greater than or equal to
- 10  $15 \text{ (J/cm}^3\text{)}^{1/2}$ .

11. The use as claimed in any one of the preceding claims, characterized in that the nonionic surfactant is chosen from:

- . polyoxyalkylenated (polyethoxyethylenated,
- 15 polyoxypropylenated or polyoxybutylenated)
- alkylphenols in which the alkyl substituent is  $C_6-C_{12}$  and containing from 5 to 25 oxyalkylene units;
- . polyoxyalkylenated  $C_8-C_{22}$  aliphatic alcohols containing from 1 to 25 oxyalkylene (oxyethylene or
- 20 oxypropylene) units;
- . products resulting from the condensation of ethylene oxide and/or propylene oxide with propylene glycol or ethylene glycol;
- . ethoxylated and/or propoxylated  $C_8-C_{18}$  fatty acids
- 25 containing from 5 to 25 ethoxylated and/or propoxylated units;
- . alkoxyated amido amines containing from 1 to 50 oxyalkylenated units;

. alkoxyated terpenic hydrocarbons such as ethoxylated and/or propoxylated  $\alpha$ - or  $\beta$ -pinenes, containing from 1 to 30 oxyethylene and/or oxypropylene units;

. alkylpolyglycosides which may be obtained by

5     condensation of glucose with primary fatty alcohols containing a C<sub>4</sub>-C<sub>20</sub> alkyl group and also an average number of glucose units of about from 0.5 to 3 per mole of alkylpolyglycoside.

10     12. The use as claimed in any one of the preceding claims, characterized in that the amount of copolymer relative to the relatively nonpolar compound or the nonionic surfactant is between 0.1% and 15% by weight and preferably between 0.5% and 10% by weight.

15     13. The use as claimed in any one of the preceding claims, characterized in that, in the case of gelation of a relatively nonpolar compound, the copolymer is combined with a nonionic or anionic surfactant.

20     14. The use as claimed in the preceding claim, characterized in that the amount of nonionic or anionic surfactant relative to the relatively nonpolar compound is between 5% and 20% by weight.

25     15. The use as claimed in any one of the preceding claims, characterized in that the copolymer is combined with a filler of lamellar structure.

16. The use as claimed in the preceding claim, characterized in that the amount of filler represents up to 20% by weight of the copolymer.

17. The use as claimed in either of claims 15 and 16, characterized in that the filler is introduced during the preparation of the copolymer and/or during the use of said copolymer.

5 18. The use as claimed in any one of claims 1 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the composition of formulations intended for cleaning metals.

10 19. The use as claimed in any one of claims 1 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the composition of detergent formulations which may be used in the industrial field.

15 20. The use as claimed in any one of claims 1 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the composition of formulations intended for stripping paints and varnishes.

20 21. The use as claimed in any one of claims 15 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the composition of formulations intended for cleaning or stripping vertical surfaces.

25 22. The use as claimed in any one of claims 1 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the composition of formulations

intended for treating plants.

23. The use as claimed in any one of claims 1 to 17, characterized in that the copolymer and the relatively nonpolar compound or the nonionic surfactant form part of the composition of formulations used in the field of printing inks.

**COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY**  
(Includes Reference to Provisional and PCT International Applications)

Attorney's Docket No.

022701-951

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name;

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

USE OF COPOLYMERS DERIVED FROM POLYAMIDES AS GELLING AGENTS FOR COMPOUNDS WITH  
LOW POLARITY

the specification of which (check only one item below):

☐ is attached hereto.

☐ was filed as United States application

Number \_\_\_\_\_  
on \_\_\_\_\_  
and was amended  
on \_\_\_\_\_ (if applicable).

☐ was filed as PCT international application

Number PCT/FR00/00687  
on 20 MARCH 2000  
and was amended  
on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 (a)-(e) of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

**PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. §119:**

COUNTRY (if PCT, indicate "PCT")	APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 35 U.S.C. §119
FRANCE	99/03,453	19 MARCH 1999	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No
			<input type="checkbox"/> Yes <input type="checkbox"/> No

I hereby claim the benefit under Title 35, United States Code § 119(e) of any United States provisional application(s) listed below.

\_\_\_\_\_  
(Application Number)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Application Number)

\_\_\_\_\_  
(Filing Date)

**COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (CONT'D)**  
(Includes Reference to Provisional and PCT International Applications)

Attorney's Docket No.

022701-951

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose to the Office all information known to me to be material to the patentability as defined in Title 37, Code of Federal Regulations §1.56, which became available between the filing date of the prior application(s) and the national or PCT international filing date of this application:

PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C. §120:

U.S. APPLICATIONS		STATUS (check one)		
U.S. APPLICATION NUMBER	U.S. FILING DATE	PATENTED	PENDING	ABANDONED
PCT APPLICATIONS DESIGNATING THE U.S.				
PCT APPLICATION NO.	PCT FILING DATE	U.S. APPLICATION NUMBERS ASSIGNED (if any)		

I hereby appoint the following attorneys and agent(s) to prosecute said application and to transact all business in the Patent and Trademark Office connected therewith and to file, prosecute and to transact all business in connection with international applications directed to said invention:

William L. Mathis	17,337	Eric H. Weisblatt	30,505	Bruce T. Wieder	33,815
Robert S. Swecker	19,885	James W. Peterson	26,057	Todd R. Walters	34,040
Platon N. Mandros	22,124	Teresa Stanek Rea	30,427	Ronni S. Jillions	31,979
Benton S. Duffett, Jr.	22,030	Robert E. Krebs	25,885	Harold R. Brown III	36,341
Norman H. Stepno	22,716	William C. Rowland	30,888	Allen R. Baum	36,086
Ronald L. Grudziecki	24,970	T. Gene Dillahunt	25,423	Brian P. O'Shaughnessy	32,747
Frederick G. Michaud, Jr.	26,003	Patrick C. Keane	32,858	Kenneth B. Leffler	36,075
Alan E. Kopecki	25,813	B. Jefferson Boggs, Jr.	32,344	Fred W. Hathaway	32,236
Regis E. Slutter	26,999	William H. Benz	25,952	Wendi L. Weinstein	34,456
Samuel C. Miller, III	27,360	Peter K. Skiff	31,917	Mary Ann Dillahunt	34,576
Robert G. Mukai	28,531	Richard J. McGrath	29,195		
George A. Hovanec, Jr.	28,223	Matthew L. Schneider	32,814		
James A. LaBarre	28,632	Michael G. Savage	32,596		
E. Joseph Gess	28,510	Gerald F. Swiss	30,113		
R. Danny Huntington	27,903	Charles F. Wieland III	33,096		



and: \_\_\_\_\_

Address all correspondence to:



**21839**

Teresa Stanek Rea  
BURNS, DOANE, SWECKER & MATHIS, L.L.P.  
P.O. Box 1404  
Alexandria, Virginia 22313-1404


Address all telephone calls to: Teresa Stanek Rea at (703) 836-6620.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

**COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (CONT'D)**  
(Includes Reference to Provisional and PCT International Applications)

Attorney's Docket No.

022701-951


FULL NAME OF SOLE OR FIRST INVENTOR Edith CANIVENC		SIGNATURE 	DATE 27/11/01
RESIDENCE 133, avenue des Freres Lumiere, F-69008, Lyon FRANCE		CITIZENSHIP FRANCE	
POST OFFICE ADDRESS 133, avenue des Freres Lumiere, F-69008, Lyon FRANCE <i>FLX</i>			
FULL NAME OF SECOND JOINT INVENTOR, IF ANY Marie-Eve PERIER		SIGNATURE	DATE
RESIDENCE 23, avenue Hector Berlioz, F-69009, Lyon FRANCE		CITIZENSHIP FRANCE	
POST OFFICE ADDRESS 23, avenue Hector Berlioz, F-69009, Lyon FRANCE			
FULL NAME OF THIRD JOINT INVENTOR, IF ANY Sylvie BODRERO		SIGNATURE	DATE
RESIDENCE 58, place des Basses Barolles, F-69230, Saint-Genis-Laval FRANCE		CITIZENSHIP FRENCH	
POST OFFICE ADDRESS 58, place des Basses Barolles, F-69230, Saint-Genis-Laval FRANCE			
FULL NAME OF FOURTH JOINT INVENTOR, IF ANY Mikel MORVAN		SIGNATURE	DATE
RESIDENCE 30-32, rue de Belfort, F-92400, Courbevoie FRANCE		CITIZENSHIP FRENCH	
POST OFFICE ADDRESS 30-32, rue de Belfort, F-92400, Courbevoie FRANCE			
FULL NAME OF FIFTH JOINT INVENTOR, IF ANY Jean-Paul LAGRANGE		SIGNATURE	DATE
RESIDENCE 24, rue G.M. Witkowski, F-69005, Lyon FRANCE		CITIZENSHIP FRENCH	
POST OFFICE ADDRESS 24, rue G.M. Witkowski, F-69005, Lyon FRANCE			
FULL NAME OF SIXTH JOINT INVENTOR, IF ANY		SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF SEVENTH JOINT INVENTOR, IF ANY		SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF EIGHTH JOINT INVENTOR, IF ANY		SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			




**COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (CONT'D)**  
(Includes Reference to Provisional and PCT International Applications)

Attorney's Docket No.

022701-951

FULL NAME OF SOLE OR FIRST INVENTOR		SIGNATURE		DATE
Edith CANIVENC				
RESIDENCE			CITIZENSHIP	
133, avenue des Freres Lumiere, F-69008, Lyon FRANCE			FRANCE	
POST OFFICE ADDRESS				
133, avenue des Freres Lumiere, F-69008, Lyon FRANCE				
FULL NAME OF SECOND JOINT INVENTOR, IF ANY		SIGNATURE		DATE
Marie-Eve PERIER				20/11/01
RESIDENCE			CITIZENSHIP	
23, avenue Hector Berlioz, F-69009, Lyon FRANCE			FRANCE	
POST OFFICE ADDRESS				
23, avenue Hector Berlioz, F-69009, Lyon FRANCE <i>FRX</i>				
FULL NAME OF THIRD JOINT INVENTOR, IF ANY		SIGNATURE		DATE
Sylvie BODRERO				
RESIDENCE			CITIZENSHIP	
58, place des Basses Barolles, F-69230, Saint-Genis-Laval FRANCE			FRENCE	
POST OFFICE ADDRESS				
58, place des Basses Barolles, F-69230, Saint-Genis-Laval FRANCE				
FULL NAME OF FOURTH JOINT INVENTOR, IF ANY		SIGNATURE		DATE
Mikel MORVAN				
RESIDENCE			CITIZENSHIP	
30-32, rue de Belfort, F-92400, Courbevoie FRANCE			FRENCH	
POST OFFICE ADDRESS				
30-32, rue de Belfort, F-92400, Courbevoie FRANCE				
FULL NAME OF FIFTH JOINT INVENTOR, IF ANY		SIGNATURE		DATE
Jean-Paul LAGRANGE				
RESIDENCE			CITIZENSHIP	
24, rue G.M. Witkowski, F-69005, Lyon FRANCE			FRENCH	
POST OFFICE ADDRESS				
24, rue G.M. Witkowski, F-69005, Lyon FRANCE				
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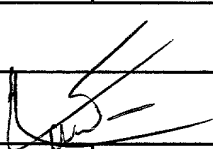
COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (CONT'D) (Includes Reference to Provisional and PCT International Applications)	Attorney's Docket No. 022701-951
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FULL NAME OF SOLE OR FIRST INVENTOR Edith CANIVENC	SIGNATURE	DATE
RESIDENCE 133, avenue des Freres Lumiere, F-69008, Lyon FRANCE	CITIZENSHIP FRANCE	
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FULL NAME OF SECOND JOINT INVENTOR, IF ANY Marie-Eve PERIER	SIGNATURE	DATE
RESIDENCE 23, avenue Hector Berlioz, F-69009, Lyon FRANCE	CITIZENSHIP FRANCE	
POST OFFICE ADDRESS 23, avenue Hector Berlioz, F-69009, Lyon FRANCE		
FULL NAME OF THIRD JOINT INVENTOR, IF ANY Sylvie BODRERO <b>Patrick BODRERO legal</b>	SIGNATURE 	DATE 5/01/2002
RESIDENCE 32, rue Charles de Gaulle, F-69340, Vaulx-en-Velin FRANCE <i>3-11</i>	CITIZENSHIP FRENCH	
POST OFFICE ADDRESS 58, place des Basses Barolles, F-69230, Saint-Genis-Laval FRANCE <i>FLX</i>		
FULL NAME OF FOURTH JOINT INVENTOR, IF ANY Mikel MORVAN	SIGNATURE	DATE
RESIDENCE 30-32, rue de Belfort, F-92400, Courbevoie FRANCE	CITIZENSHIP FRENCH	
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RESIDENCE	CITIZENSHIP	
POST OFFICE ADDRESS		

**COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (CONT'D)**  
(Includes Reference to Provisional and PCT International Applications)

Attorney's Docket No.

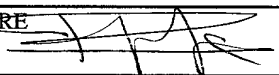
022701-951

FULL NAME OF SOLE OR FIRST INVENTOR		SIGNATURE	DATE
Edith CANIVENC			
RESIDENCE		CITIZENSHIP	
133, avenue des Freres Lumiere, F-69008, Lyon FRANCE		FRANCE	
POST OFFICE ADDRESS			
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FULL NAME OF SECOND JOINT INVENTOR, IF ANY		SIGNATURE	DATE
Marie-Eve PERIER			
RESIDENCE		CITIZENSHIP	
23, avenue Hector Berlioz, F-69009, Lyon FRANCE		FRANCE	
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FULL NAME OF THIRD JOINT INVENTOR, IF ANY		SIGNATURE	DATE
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RESIDENCE		CITIZENSHIP	
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**COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (CONT'D)**  
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Attorney's Docket No.

022701-951

FULL NAME OF SOLE OR FIRST INVENTOR		SIGNATURE		DATE
Edith CANIVENC				
RESIDENCE		CITIZENSHIP		
133, avenue des Freres Lumiere, F-69008, Lyon FRANCE		FRANCE		
POST OFFICE ADDRESS				
133, avenue des Freres Lumiere, F-69008, Lyon FRANCE				
FULL NAME OF SECOND JOINT INVENTOR, IF ANY		SIGNATURE		DATE
Marie-Eve PERIER				
RESIDENCE		CITIZENSHIP		
23, avenue Hector Berlioz, F-69009, Lyon FRANCE		FRANCE		
POST OFFICE ADDRESS				
23, avenue Hector Berlioz, F-69009, Lyon FRANCE				
FULL NAME OF THIRD JOINT INVENTOR, IF ANY		SIGNATURE		DATE
Sylvie BODRERO				
RESIDENCE		CITIZENSHIP		
58, place des Basses Barolles, F-69230, Saint-Genis-Laval FRANCE		FRENCH		
POST OFFICE ADDRESS				
58, place des Basses Barolles, F-69230, Saint-Genis-Laval FRANCE				
FULL NAME OF FOURTH JOINT INVENTOR, IF ANY		SIGNATURE		DATE
Mikel MORVAN				
RESIDENCE		CITIZENSHIP		
30-32, rue de Belfort, F-92400, Courbevoie FRANCE		FRENCH		
POST OFFICE ADDRESS				
30-32, rue de Belfort, F-92400, Courbevoie FRANCE				
FULL NAME OF FIFTH JOINT INVENTOR, IF ANY		SIGNATURE		DATE
Jean-Paul LAGRANGE				22/11/2001
RESIDENCE		CITIZENSHIP		
24, rue G.M. Witkowski, F-69005, Lyon FRANCE		FRENCH		
POST OFFICE ADDRESS				
24, rue G.M. Witkowski, F-69005, Lyon FRANCE				
FULL NAME OF SIXTH JOINT INVENTOR, IF ANY		SIGNATURE		DATE
RESIDENCE		CITIZENSHIP		
POST OFFICE ADDRESS				
FULL NAME OF SEVENTH JOINT INVENTOR, IF ANY		SIGNATURE		DATE
RESIDENCE		CITIZENSHIP		
POST OFFICE ADDRESS				
FULL NAME OF EIGHTH JOINT INVENTOR, IF ANY		SIGNATURE		DATE
RESIDENCE		CITIZENSHIP		
POST OFFICE ADDRESS				